Alternating spin-1/spin-$\frac{1}{2}$ sites in a diamond lattice: Ground state and excitations

Swapan K. Pati
Theoretical Sciences Unit, Jawharlal Nehru Center for Advanced Scientific Research, Jakkur Campus, Bangalore 560 064, India
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We consider a diamond lattice with spin-1 and spin-$\frac{1}{2}$ sites in alternating positions. The system is shown to be mapped on to a spin-1 sawtooth lattice for a range of parameter values. With varying strengths of interactions between spin sites, we find a number of phases, including, gapped spin-1, dimerized, and gapless spin-1 phases. In the gapped phase, there exist two special lines, namely, disorder and the Lifshitz lines, which differentiate spin ordering from the Néel type to spiral type, in real and in the momentum space, respectively. The dimerized phase, which ends at disorder line, is gapless in this case and corresponds to nearest-neighbor spin-$\frac{1}{2}$ sites forming alternate singlet and triplet.

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Recently the studies of insulating magnets with geometrical frustrations have generated huge interest because of their unique low-energy characteristics.[1] In general, frustrations arise due to competing interactions. After Haldane's conjecture,[2] many theories have been developed to distinguish the effects of frustrations between integer and half-odd-integer spin chains.[3–5] Parallely, frustrated spin-$\frac{1}{2}$ and spin-1 chains have been realized experimentally.[6]

There exist a large number of spin systems where frustrations arise due to the topological arrangements of metal ions. One of the most studied systems in recent years is the two-dimensional Kagome antiferromagnets, in which frustrations lead to a very complex excitations spectrum.[7] However, frustrated systems, where two or more different metal ions are present in the unit cell, have received very little theoretical attention. The magnetic interactions between two nonequivalent magnetic centers mediated by topological frustrations may lead to situations that may not be encountered with species containing a unique kind of magnetic ions.[8]

A large number of such low-dimensional heteropolymetallic frustrated systems have been synthesized experimentally. For example, there exist planar tetranuclear complexes with topological frustrations. The general structures of these complexes can vary from butterfly, pyramidal to lozenge, with two different magnetic ions.[9] More specifically, the series $Ba_2CaMM'_2F_{14}$, where M and M' are the transition metal ions, form chains of lozenges and the chains are well seperated by Ca^{+2} ions.[10] In each chain, M' can interact with the two nearest-neighbor M sites, while each M can interact with four M's. These systems have been found to exhibit a number of different magnetic structures, which are extremely sensitive to the magnitudes of the competing antiferromagnetic interactions. This is an example of frustrated ferrimagnet and such a structure when connected to form an extended network in one dimension is the focus of our paper.

The structure that we have considered is shown in Fig. 1(a), with site spins $S=1$ and $s=1/2$ residing in four corners of a diamond structure. Similar structure has been considered for $S=s=1/2$[11] and with different spins.[13] On the other hand, Sen *et al.*[14] have studied a spin-$\frac{1}{2}$ sawtooth lattice, structure of which is shown in Fig. 1(b). This structure resembles a spin chain with nearest- and next-nearest neighbor interactions with every alternate exchange interaction between next-nearest neighbors being missing.

The Hamiltonian for our system in Fig. 1(a) can be written as

$$H=\sum_{i=1}^{N} J(S_{1,i}+S_{1,i+1})(s_{2,i}+s_{3,i})+J'S_{1,i}S_{1,i+1}+J''s_{2,i}s_{3,i}, \tag{1}$$

where the sum runs over the number of unit cells (each unit cell contains three spin sites). $S_{1,i}$ and $s_{2,i}=s_{3,i}$ correspond to the site spin with magnitude $S=1$ and $s=1/2$ in the i^{th} unit cell. We consider a periodic boundary condition, with $S_{1,N+1}=S_{1,1}$, for system with N unit cells. This Hamiltonian commutes with the operator $(s_{2,i}+s_{3,i})$, for every unit cell i.

We have implemented the density-matrix renormalization group (DMRG) method,[15] which is considered to be a highly accurate method for interacting systems in low dimensions.[15,5] The density-matrix cutoff from $m=150$ to $m=200$ have been used throughout, and the results have been extensively verified with the exact diagonalization calculations for small system sizes.[16] We have calculated the equal time spin-spin correlations and the energy in the ground state. We have also obtained excitation gaps with a varying range of exchange parameters. In some cases, we have computed dispersion spectrum by exact diagonalization method for small system sizes.

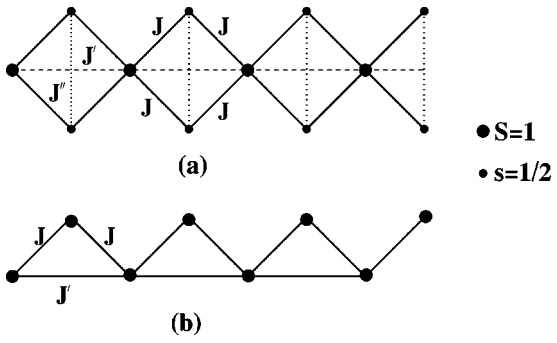

FIG. 1. Schematic picture of the arrangement of (a) spin-1 and spin-1/2 sites in a diamond lattice and (b) spin-1 sites in a sawtooth lattice.

Let us first discuss the results considering $J''=0$. Unlike the case as in Ref. 11, with $J'=0$, the system has a ground state that is a singlet. It is easy to see that spins are exactly compensated in each unit cell. We find that even when J' is quite large, the ground state remains a singlet. There is a quantum phase transition as we increase J' from zero to a higher value. The spin-1 sites remain aligned parallely at small values of J' but becomes strongly antiferromagnetic as we increase J'. However, the operator $(s_{2,i}+s_{3,i})$ remains constant and it is a triplet in each unit cell i for all J' values. By considering each of these spin-$\frac{1}{2}$ pairs as a spin 1, the system can be mapped to a sawtooth lattice with spin-1 sites at each vertex [see Fig. 1(b)]. The sawtooth lattice with various site spins have been synthesized in recent years.[12] Our lattice structure turns out to be exactly the spin-1 chain with nearest- and next-nearest neighbor interactions with every alternate next-neighbor interaction being missing.

The spin-$\frac{1}{2}$ analog of this sawtooth lattice has been studied in detail in Ref. 14. Elementary excitations in these class of systems are found to consist of kink-antikink pairs, while the ground state is doubly degenerate. However, its spin-1 analog is completely different. At $J'=0$, the system is a spin-1 chain with nearest-neighbor exchange. It is well established by now that the spin-1 chain has a finite and short correlation length of $\xi=6.03$, a Haldane gap $\Delta=0.41050$ and is characterized by a *hidden* $Z_2\times Z_2$ symmetry breaking order.[2,17,19] The ground-state energy, the Haldane gap, and the correlation length have been found to be correct to numerical accuracy by the DMRG method.[19]

As we increase J' from zero, the system remains in the spin-1 gapped phase, until $J'=1.0\pm0.05$. Within this phase, we have found two special points. One is the disorder point, and another is the Lifshitz point. These points have been observed for the spin-1 chains with nearest and next-nearest neighbor antiferromagnetic exchanges.[4] A disorder point is a point where the system changes from one broken symmetry phase to another at low temperature. In general, in spin systems it is defined as a point where the system changes smoothly from antiferromagnetically ordered phase to a spiral phase with incommensurate wave vector. Therefore, in the disorder phase, the correlation lengths would be short ranged of the type as in a spin-1 ordered phase. Classical frustrated spin chain shows similar transition from the Néel ordered state to a spiral ordered state.[20] In the analogy with the general bilinear-biquadratic spin chain [Affleck, Kennedy, Lieb, and Tasaki (AKLT) model][17] in its classical limit, we can fit the static correlation functions to a two-dimensional Ornstein-Zernike correlation function

$$\langle S_iS_j\rangle \propto \frac{\exp(-|i-j|/\xi)}{\sqrt{|i-j|}}, \tag{2}$$

where $|i-j|$ is the distance between the sites i and j, and ξ is the correlation length. The disorder phase in this case, as first identified in the AKLT model, is a quantum remnant of the Néel order to spiral order transition in the classical frustrated chain at low-temperature. In Fig. 2, we plot the function $\ln(|\langle S_iS_j\rangle|\sqrt{|i-j|})$ as a function of $|i-j|$. As can be seen, the function shows incommensurate behavior at the disorder point which is at $J'=0.5$. Below this point, the data fit to a straight-line equation with numerical accuracy.

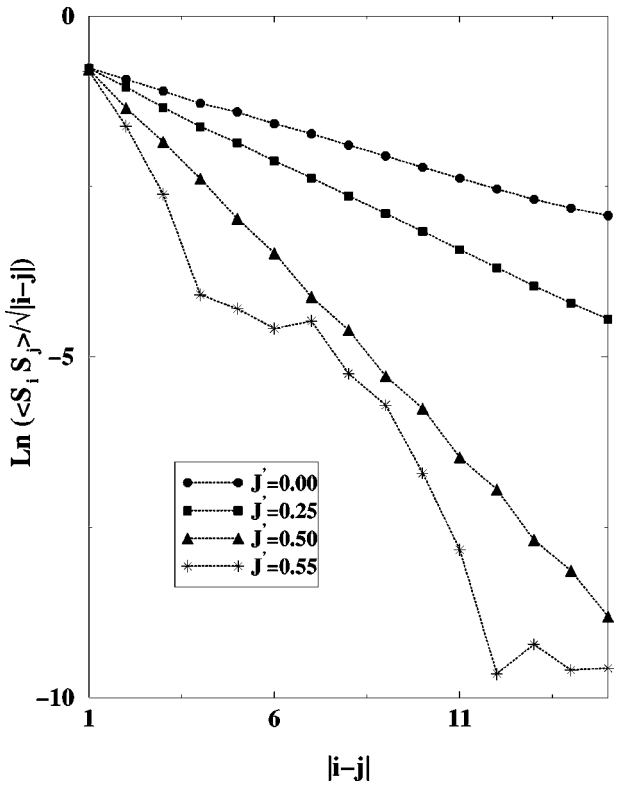

FIG. 2. Log of $(|\langle S_iS_j\rangle|\sqrt{|i-j|})$ as a function of the distance $|i-j|$, for a number of J' values.

It is to be noted that in the disorder phase, the momentum space spin-correlation functions peak at $q_{max}=0$. However, as the correlations are short ranged, the momentum space correlation functions slowly start moving from $q_{max}=0$ to some general $q_{max}>0$. This occurs at some point in the incommensurate-correlation region, away from the disorder point. This is the other special point, called the Lifshitz point. The structure factor $S(q)$ which is defined as

$$S(q)=\sum_{|i-j|}\exp(iq|i-j|)\langle S_iS_j\rangle \tag{3}$$

develops peaks in an incommensurate values of wavenumber q_{max}. We have plotted $S(q)$ as a function of q in Fig. 3. Below $J'=0.75$, the structure factor peaks at $q_{max}=0$ or $q_{max}=2\pi$. However above this point, the q_{max} changes to incommensurate values, $0<q_{max}<\pi/2$. The Lifshitz point has been predicted to be associated with a doubly degenerate structure of the solitonic excited states. Note that only this point can be rationalized experimentally, since the experimental structure factor is associated with the wavenumber.

Until J' reaches the Lifshitz point, the two-spin correlations between the nearest-neighbor spin-1 sites remain ferromagnetic in character. However, as we increase J', this two-point correlation becomes very small. Note that even zero correlation between the spin-1 sites will not leave behind any free spin. Further increase of J' develops in antiferromagnetic character between the nearest-neighbor spin-1 sites.

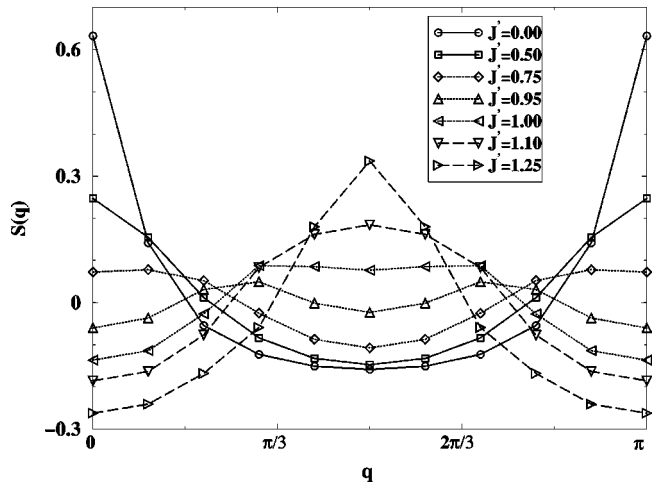

FIG. 3. Structure factor $S(q)$ versus the wave vector, q, for a number of J' values.

Surprisingly, this occurs precisely at $J'=1\pm0.05$. It can also be observed from Fig. 3: the structure factor develops peak at $q_{max}=\pi$ close to $J'=1$.

In the limit, $J'\to\infty$, the frustrated spin-1 system will be decomposed into a next-nearest-neighbor spin-1 chain (with every alternate one missing), and a free spin-1 in each unit cell. The free spin will give rise to gapless excitations. For this to happen, the spin-1 neighbors should develop antiferromagnetic character. As discussed above, the antiferromagnetic correlations between spin-1 neighbors appear at $J'=1\pm.05$, which indicates the phase transition. This transition point can be characterized as the point where the system goes from the standard AKLT phase to a next-nearest neighbor (with every alternate one missing) gapless AKLT phase.

The ground-state energy changes its slope as a function of J', above $J'=1.0$ [see Fig. 4(a)]. We have also obtained the lowest-energy gap as a function of J', which is shown in Fig. 4(b). The energy gap starts from the Haldane gap value (0.401) when $J'=0$, goes through a maximum near the Lifshitz point, and then decreases smoothly to zero as the phase changes. The standard spin-1 Haldane phase is characterized by a so-called string order parameter, which measures the *hidden* order due to the symmetry breaking,[18] and is defined as

$$O^z(m-n)=\left\langle S_m^z\left(\exp\sum_{k=m+1}^{n} i\pi S_k^z\right)S_n^z\right\rangle. \qquad (4)$$

We have computed the string order parameter $O^z=\lim_{|m-n|\to\infty}O^z(m-n)$ and found that it is nonzero throughout the gapped phase in the J' line, and it drops to zero discontinuously at the phase transition, when the systems enter into the gapless phase. This is shown in Fig. 4(c). Also note that at $J'>1$, the correlation function between the spin-1 and spin-1/2 neighbors become almost zero, confirming that the spin-1/2 nearest-neighbors that form triplets, are free in nature.

We have also calculated the lowest spin excitation dispersion curve for a number of J' values for a system size of $N=18$. The spin-1 solitonic excitation has a minimum at $q=0$ or 2π. However, as J' increases, the excitation minimum shifts to $q=\pi/3$ or $2\pi/3$ close to the disorder point. Further increase of J' shows that the minimum is at $q=\pi$. It is difficult to determine this point, as the system size studied is very small. However, a cautious estimation puts this close to the Lifshitz point. Eventually as discussed above, this excitation gap at $q=\pi$ vanishes in the long chain limit, as the phase changes to a gapless phase.

So far we have discussed the results with $J''=0$. However, with nonzero J'', some additional interesting features emerge. The ground state remains a singlet for all J'' values, but dimerization sets in at various points in the $J'-J''$ phase diagram. For small J'', all $(s_{2,i}+s_{3,i}), i=1,2,\ldots,$ remain triplets as the case with $J''=0$. However, as we increase J'' from 0 $(s_{2,i}+s_{3,i})$ with alternate i^{th} unit cell, form singlet combinations. This situation can be visualized as if the interactions between spin-1 sites are mediated by the alternating

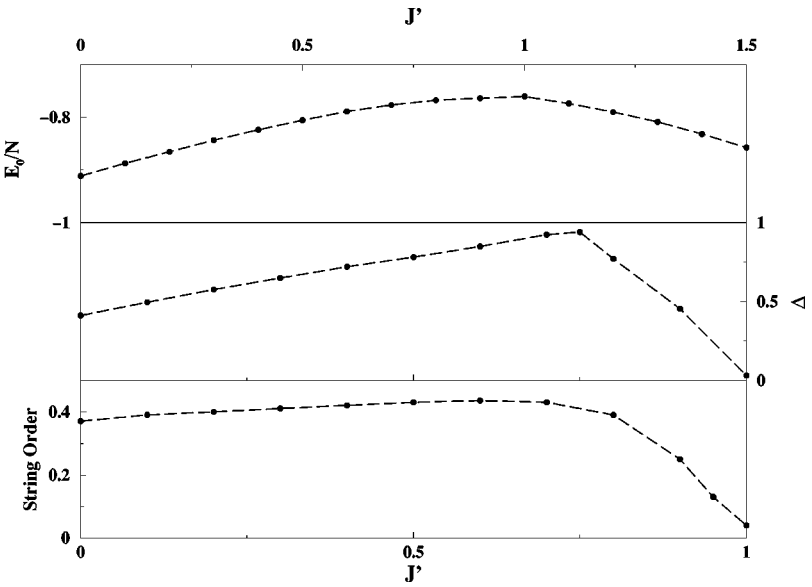

FIG. 4. (a) Ground-state energy (E_0/N), (b) lowest spin excitation gap (Δ), and (c) string order parameter (O^z), as a function of J'.

spin-1 and spin-0 sites. In other words, the spin-1 sites become decoupled, with maximum isolated segment length being 3 sites. To give an estimate, this situation arises at $J''=2.7$, when the spin 1 sites are not connected ($J'=0$). The chain becomes decoupled with a spin-1 dimer and a free spin-1 in every alternate unit cell. However, if J'' is further increased, at $J''=3$, all the $(s_{2,i}+s_{3,i}), i=1,2,\ldots$, form singlets. This limit results in a chain with completely decoupled free spin-1 sites. It is interesting to note that, the dimerized phase discussed above is gapless in this case due to free spins.

At nonzero J' values, this dimerized ground state with alternate singlet and triplet spin-1/2 combinations remains unaffected as long as $J'<0.5$. At higher J' values, the $(s_{2,i}+s_{3,i}), i=1,2,\ldots$ pairs forming singlets at alternate i^{th} unit cell arises at smaller J'' values. However, with nonzero J' values, the physics remain very similar to what is observed at $J'=0$, unless $J'<0.5$. For $J'\geqslant 0.5$, the alternate singlet/triplet in $(s_{2,i}+s_{3,i}), i=1,2,\ldots$ do not appear, rather with increase in J'', the nondegenerate ground state becomes infinitely degenerate after certain J'' values. This transition corresponds to the situation where all $(s_{2,i}+s_{3,i}), i=1,2,\ldots$ triplets, transform to singlets at some critical J'', for every $J'\geqslant 0.5$ values. We have shown it in the $J'-J''$ phase digram (line C' in Fig. 5).

In the gapped phase, both, the disorder and the Lifshitz points, extend to nonzero J'' to form two special lines, with the same features, as discussed for the case with $J''=0$. However, these two lines end at the J'' values where $(s_{2,i}+s_{3,i}), i=1,2,\ldots$, form singlets, namely, where the gapless phase appears (broken C' line in Fig. 5). It is interesting to note that the ground-state dimerized phase ends as the J' hits the disorder line.

As shown in the phase diagram (Fig. 5), there are three gapless phases. For $J''=0$, as discussed earlier, the gapless phase arises because of the large number of free spin-1 sites (spin-$\frac{1}{2}$ sites forming triplets) at $J'=1$ in a perfectly one-dimensional spin-1 chain background. However, with $J''\neq 0$, the gapless phase is due to the formation of singlets between the nearest-neighbor spin-$\frac{1}{2}$ sites, with free spin 1 in each unit cell, or in every alternate unit cell. While the shaded part in the phase diagram, which is a dimerized phase, is gapless in this case due to nearest-neighbor spin-$\frac{1}{2}$ sites forming alternate singlet and triplet, the phase with high J'' value is gapless because all the spin-1 sites become free, with all nearest-neighbor spin-$\frac{1}{2}$ sites forming singlet combinations. Moreover, for $J''=0$, all $(s_{2,i}+s_{3,i}), i=1,2,\ldots$ remain triplets. Thus, the $J''\neq 0$ gapless phase can not extend

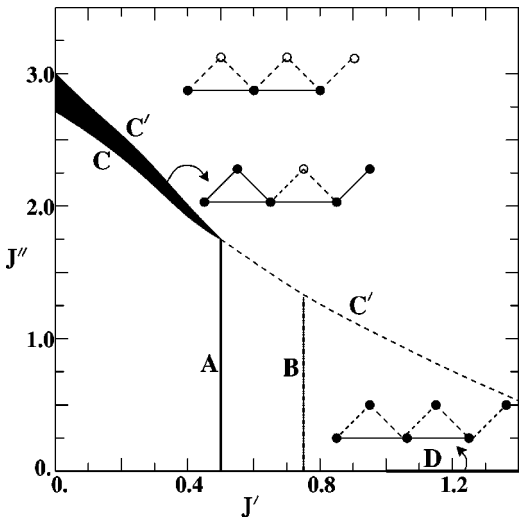

FIG. 5. Phase diagram of the model in Hamiltonian (1) in the J', J'' parameter space. Lines *A* and *B* are the disorder and Lifshitz lines, respectively. Line *C* corresponds to alternating singlet and triplet combinations, while line C' represents all singlet combinations, by the nearest-neighbor spin-$\frac{1}{2}$ sites. The shaded region is the dimerized phase and line *D* is the gapless spin-1 phase. All three gapless phases are schematically drawn for clarity, where the closed (open) circles are for spin-1 (spin-0) sites, and the solid and broken lines represent strong and weak interactions, respectively. All the phase boundaries (*C*, C', and *D* lines) are obtained numerically with error bars ± 0.05 in both the J' and J'' energy scales. The line C' ends at $(J',J'')=(1.5,0.1)$, which is not shown.

to join the $J''=0$ gapless phase. Numerically, we find that the $J''\neq 0$ gapless line (broken C' line in the phase diagram) approaches J' line (with $J''=0$) as we increase J'. However, this line ends abruptly at around $(J',J'')=(1.5,0.1)$. Below $J''=0.1$, the spin gap is zero, as it corresponds to the next-nearest-neighbor (with every alternate one missing) AKLT phase with free spin-1 sites. The main point is that in the J' line (with $J''=0$) gapless phase, only a small value of J'' can bring about singlet formation between the nearest-neighbor spin-$\frac{1}{2}$ sites, with an infinitely degenerate ground state.

To conclude, we have studied a one-dimensional diamond lattice of spin-1 and spin-$\frac{1}{2}$ sites in the alternating sites. The system is equivalent to a spin-1 sawtooth lattice, even when there are very strong exchanges between the spin-1 sites. There exist a number of low-temperature phases: the gapped nearest-neighbor AKLT phase and several gapless spin-1 phases with infinite ground-state degeneracy. In the gapped region, we find two special lines, which differentiate spin ordering from the Néel type to the spiral type.

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